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ESI: A free-standing, flexible lithium-ion anode formed from an air-dried slurry cast of high tap density SnO_2, CMC polymer binder and Super-P Li

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A free-standing electrode film composed of high tap density SnO_2 particles and carboxymethyl cellulose binder with Super-P Li (SP-Li) conductive carbon was formed from an aqueous slurry cast by doctor-blading. Upon air-drying, the free-standing film spontaneously evolved *via* delamination from the substrate as the slurry solvent evaporated. The electrodes cut from the free-standing film were ~5 μm thick with a SnO_2 loading of ~0.5 mg cm^{-2}. The films were found to be easily handled, flexed and folded. For evaluation of the durability of the free-standing films, the tensile strength and elongation at break were measured: 13 MPa and 1.7%. The robustness of the electrically conductive network was measured with a four-point probe: the initial electrical resistivity of the film (0.6 Ω-cm) was observed to increase by 6% after folding, applying pressure to the crease and unfolding. When tested in a coin cell, the electrode cycled stably with near 100% coulombic efficiency at up to 2*C* and without capacity fade for 100 cycles at 1*C*. To adjust the areal capacity of the cell, multiple free-standing films could be stacked. An electrode formed from several stacked films with an active material mass loading of greater than 4 mg cm^{-2} was found to cycle stably at 2.6 mAh cm^{-2} tested at 0.33 mA cm^{-2} current density. For evaluating cycling performance of the electrode while flexed, an electrode was placed in a once-folded pouch cell for testing at 1*C* and cycled stably for 20 cycles before slight capacity fade was observed. For free-standing electrodes, 1D or 2D carbons such as carbon nanotubes (CNT) or graphene are commonly used to provide both mechanical strength and electrical conductivity. Here, CNT were substituted for the SP-Li and similar free-standing films were made and compared. With CNT, the electrode strength at break as well as the electronic conductivity increased, but, despite this, the cycling performance of the electrodes made using the low-cost SP-Li carbon exceeds that of the electrodes made with orders-of-magnitude more expensive 1D carbon.

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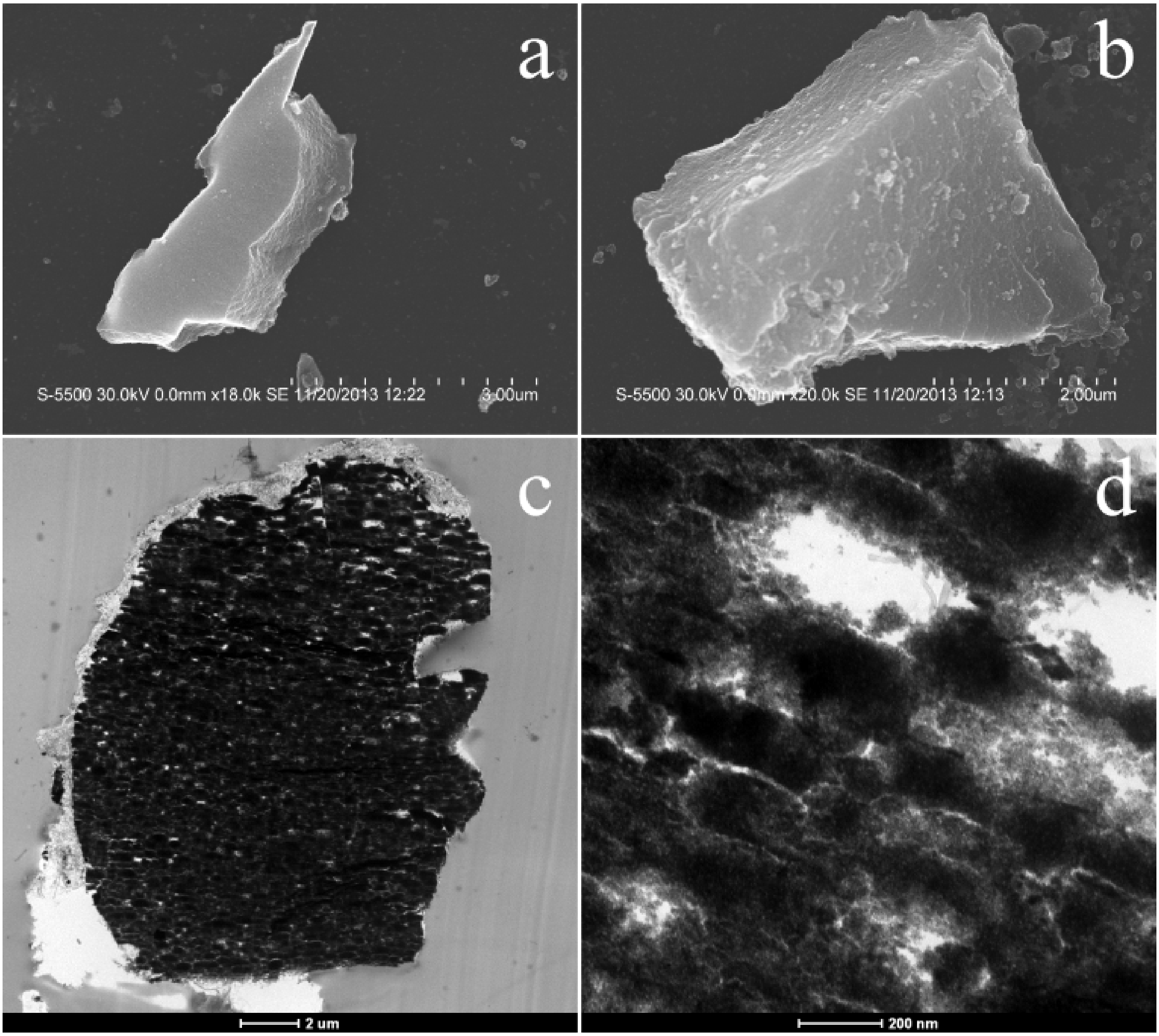
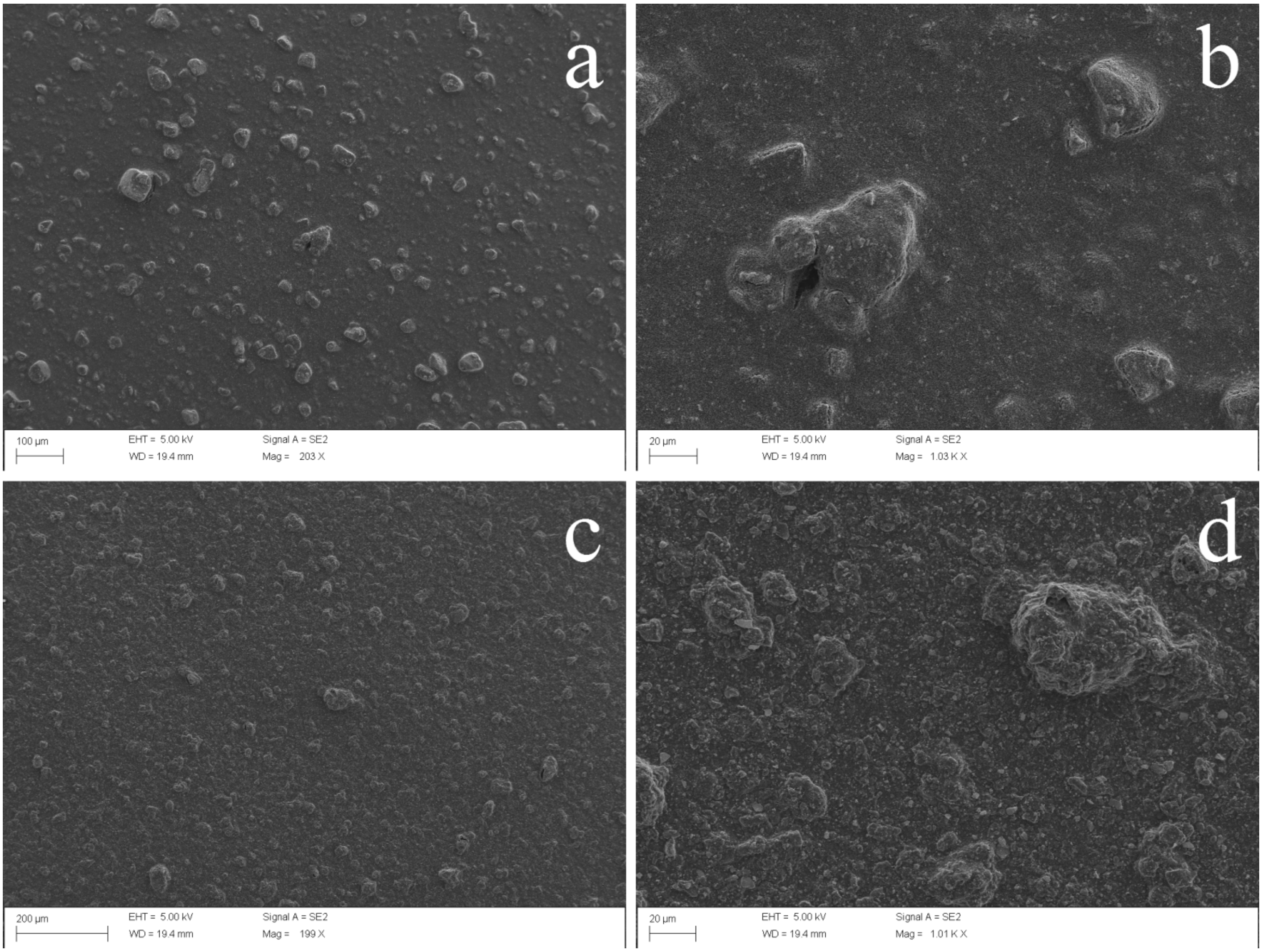
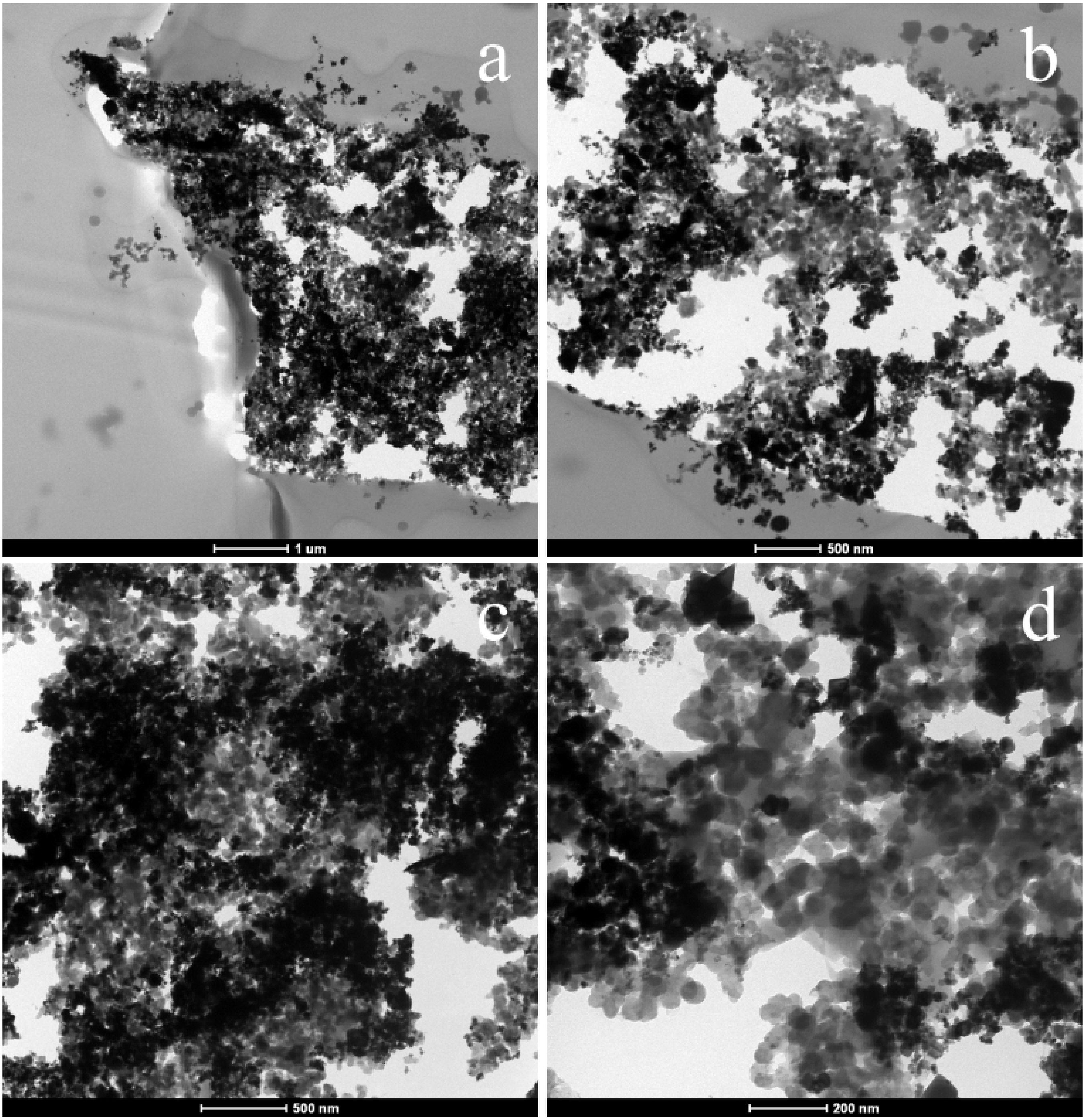


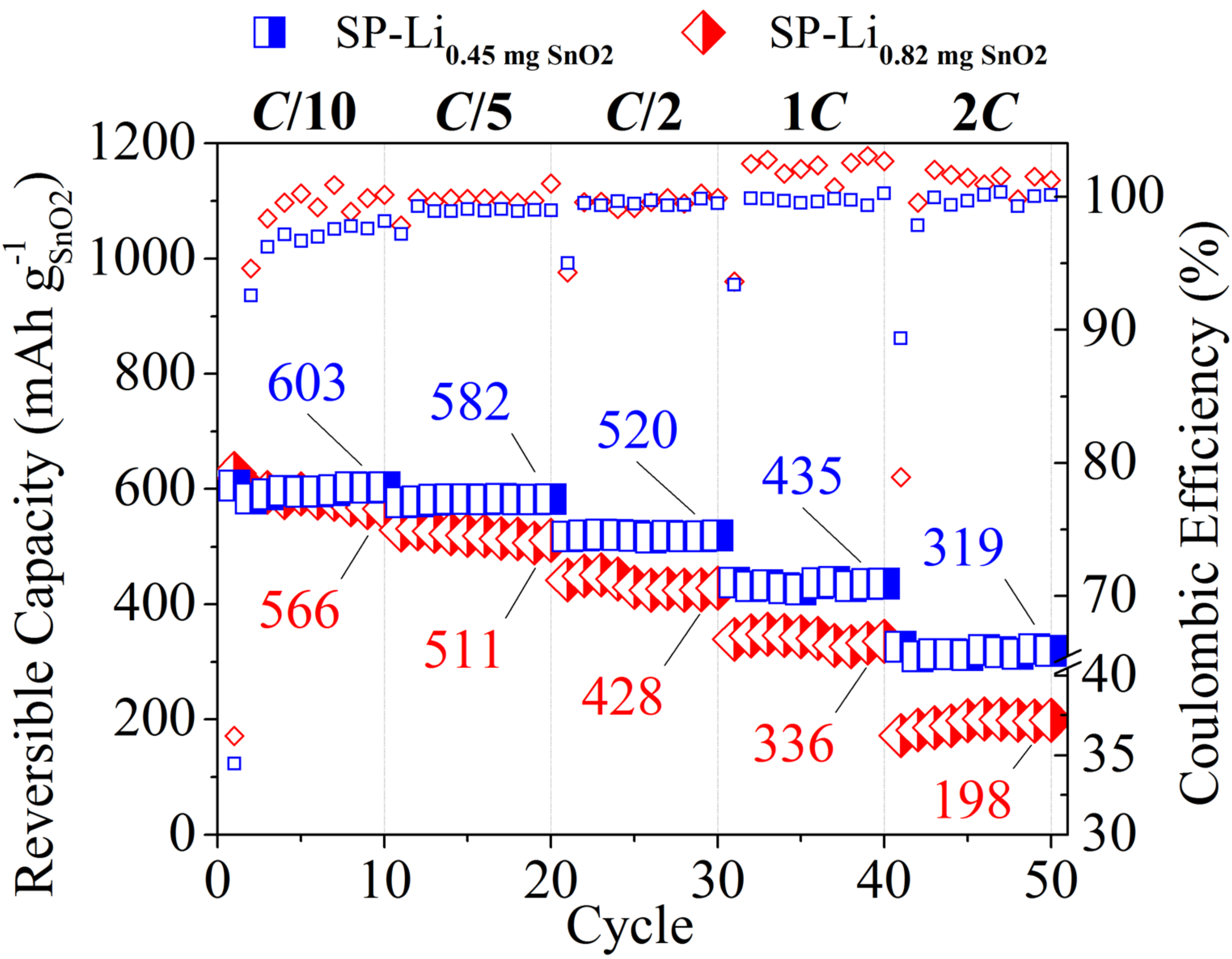
S Figure 1. SEM of SnO_2 particles (a) and (b). Cross-section TEM of particles in pristine film showing that the micron sized particle is the result of the sintering of nanoscale particles (c) and (d).



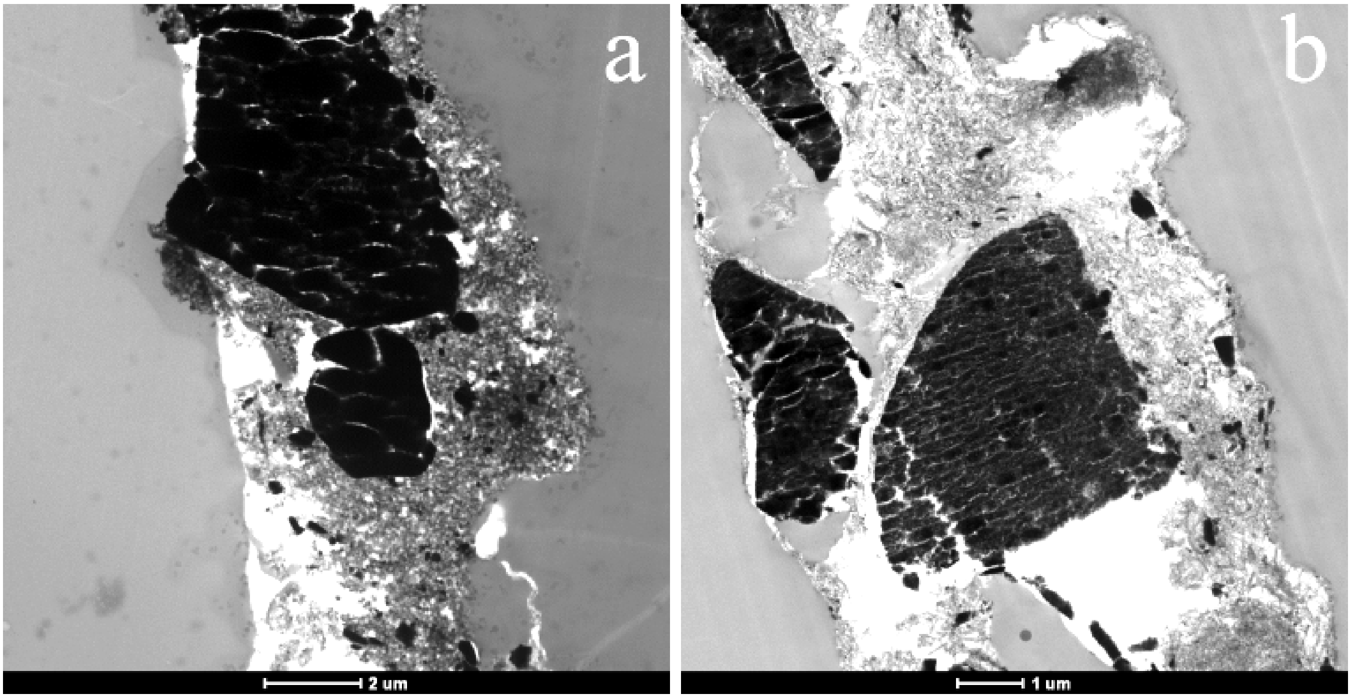
S Figure 2. SEM images at two levels of magnification showing uncycled electrodes in top down orientation featuring the surface of flexible electrode made using SP-Li conductive additive (a) – (b) and (c) – (d) with CNT. The large bulges in the film are the SnO_2 particles.



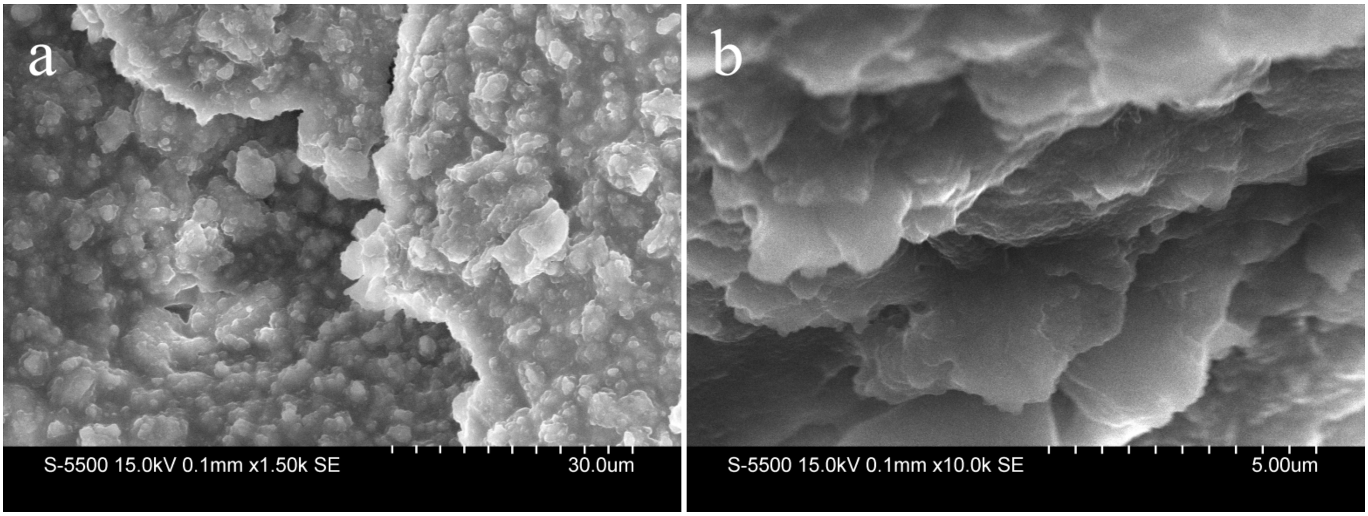
S Figure 3. TEM image of cross-section created by embedding electrode in resin and using an ultramicrotome to cut thin sections. This flexible electrode was created using low tap density, nanosized SnO_2 particles (*Sigma*, <100 nm). (a) – (d) Electrode shown at increasing magnification.



S Figure 4. C-rate test for electrode made with high tap density SnO_2 and SP-Li showing the effect of mass loading. The increase in mass loading results in higher resistance and lower capacities at faster rates.



S Figure 5. TEM, cross-sections of uncycled electrode films made with high tap density SnO_2 and (a) SP-Li or (b) CNT conductive additive.



S Figure 6. SEM images of cycled stacked anode (made with nine stacked SP-Li films) in the discharged state after 101 cycles at variable rates (see Figure 4). The SEI coats the film surface. In (a) three films are visible at different heights and (b) is zoomed in on the edge of one of the films.

Citations

1. Kim, C.; Noh, M.; Choi, M.; Cho, J.; Park, B., Critical Size of a Nano SnO_2 Electrode for Li-Secondary Battery. *Chemistry of Materials* **2005,** *17* (12), 3297-3301. http://dx.doi.org/10.1021/cm048003o